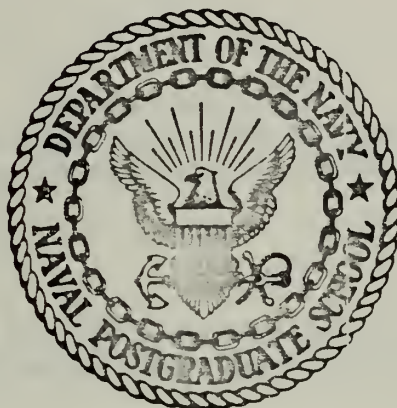


COLLISIONAL EXCITATION FOR THE FIRST
NEGATIVE AND SECOND POSITIVE SYSTEMS
OF NITROGEN

Donald Lee Cover

United States Naval Postgraduate School



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COLLISIONAL EXCITATION RATES FOR THE FIRST
NEGATIVE AND SECOND POSITIVE SYSTEMS OF NITROGEN

by

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Thesis Advisor:

E.A. Milne

September 1971

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Collisional Excitation Rates for the First
Negative and Second Positive Systems of Nitrogen

by

Donald Lee Cover
Lieutenant Commander, United States Navy
B.S., Naval Postgraduate School, 1971

Submitted in partial fulfillment of the
requirements for the degree of

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from the
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ABSTRACT

Nitrogen and nitrogen - argon gas mixtures were excited by 1.5 MeV protons from a Van De Graaff generator. Intensity measurements were made for the first negative ($\lambda = 3914.4\text{\AA}$) and the second positive ($\lambda = 3371.3\text{\AA}$) transitions of molecular nitrogen. A determination of the excitation coefficient (C), which is a function of the reaction rate coefficient and excitation cross section, was made for the nitrogen first negative and second positive transitions. The excitation coefficient (C) is a constant for each transition and contains all non-radiative energy transfer constants of the general rate equation. For the first negative transition the value of excitation coefficient was found to be $.256 \pm .046$ and for the second positive transition it was found to be $2.25 \pm .12$.

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I. INTRODUCTION

This work is a continuation of the study of the reaction rates and cross sections for the $(N_2)^*$ and $(N_2^+)^*$ bands [1] and the investigation of the quenching effect of carbon dioxide (CO_2) on the nitrogen gas system [2]. In this work the effects of Argon on the nitrogen gas system were investigated. A constant pressure of N_2 gas was maintained in the interaction chamber under bombardment by accelerated protons while increments of Argon gas were added. Intensity measurements were made at each increment up to 700 Torr. From a plot of relative intensity versus pressure a determination was made of the excitation coefficient constant which is a function of the reaction rate coefficient and excitation cross section for the nitrogen first negative transition ($B^2\Sigma_u^4 \rightarrow X^2\Sigma_g$) wavelength ($\lambda = 3914.8\text{\AA}$) and the second positive transition ($C^3\Pi_4 \rightarrow B^3\Pi_g$) ($\lambda = 3371.3\text{\AA}$).

II. EXPERIMENTAL PROCEDURE

A 2.5 MeV Van De Graaff was utilized to provide a constant energy proton beam for bombardment of the target gases in the interaction chamber. The system configuration is shown in Figure 1 and Figure 2.

The interaction chamber was a pyrex glass Tee which contained a Faraday cup for beam current collection. The beam current was integrated by an Eldorado Electronics current integrator, Model CI-110, which provided a continuous indication of beam current and total charge received at the Faraday cup. The interaction chamber connected to a liquid nitrogen trapped oil diffusion vacuum system which was capable of evacuating the chamber to 4×10^7 torr.

A gas manifold mounted on the top of the interaction chamber was used to admit research grade gases while the chamber pressure was monitored on two Wallace and Tiernan pressure gauges. One gauge had a scale of 0-50 torr with an error of $\pm .02$ torr and a second gauge with a scale of 0-800 torr with an error of ± 5 torr were used to obtain pressure measurements during the experimental runs.

A 15 cm focal length fused quartz lens located 90° from the beam axis was focused on the proton beam just as it passed through the window of the interaction chamber and focused the reaction spectrum through a mechanical chopper onto the entrance slits of the Jerral Ash monochrometer. The resolving power of the monochrometer was 10^8 about the central wavelength. The monochrometer was adjusted to select the desired wavelength which then passed

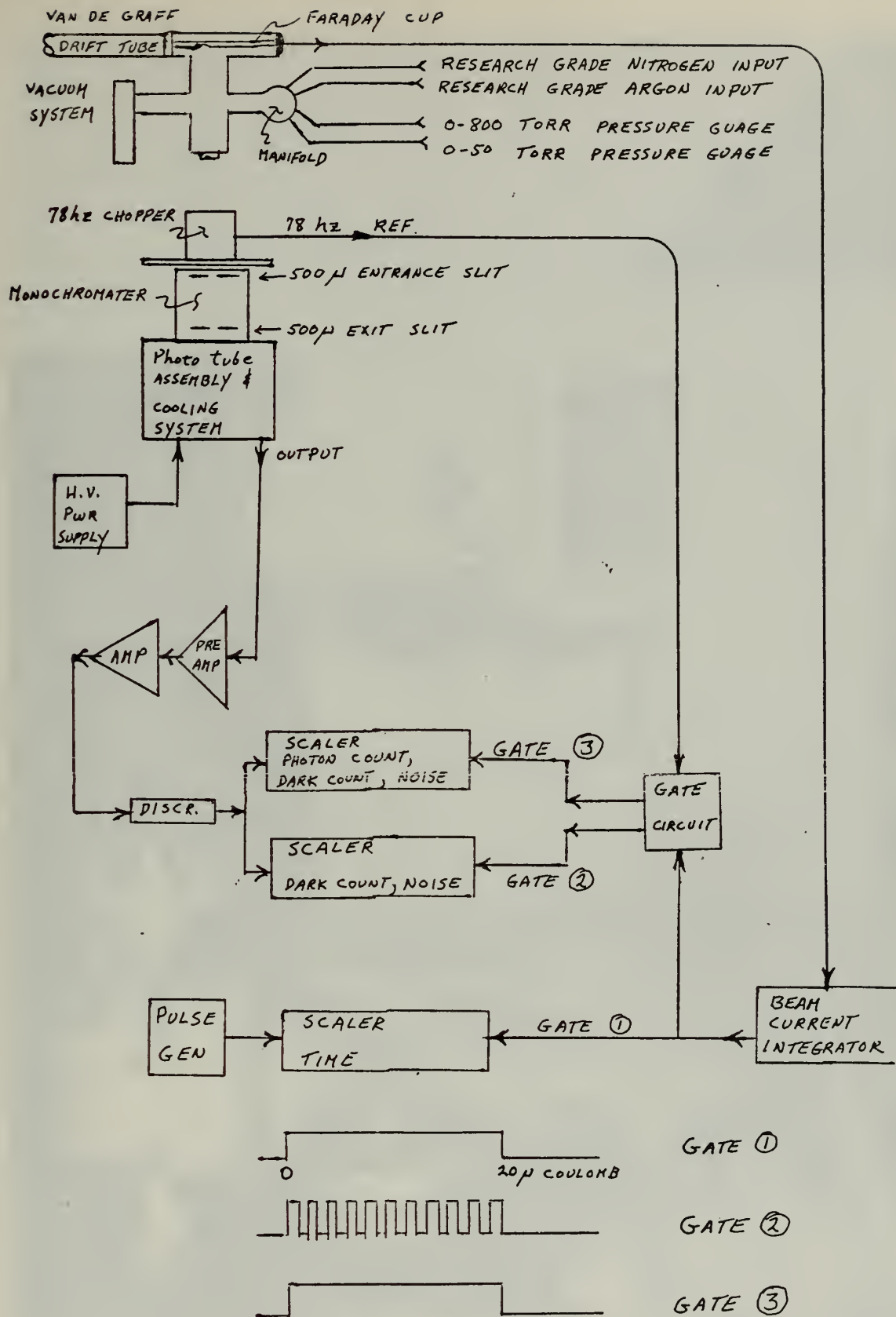


Figure 1. Instrumentation System Block Diagram and Timing.

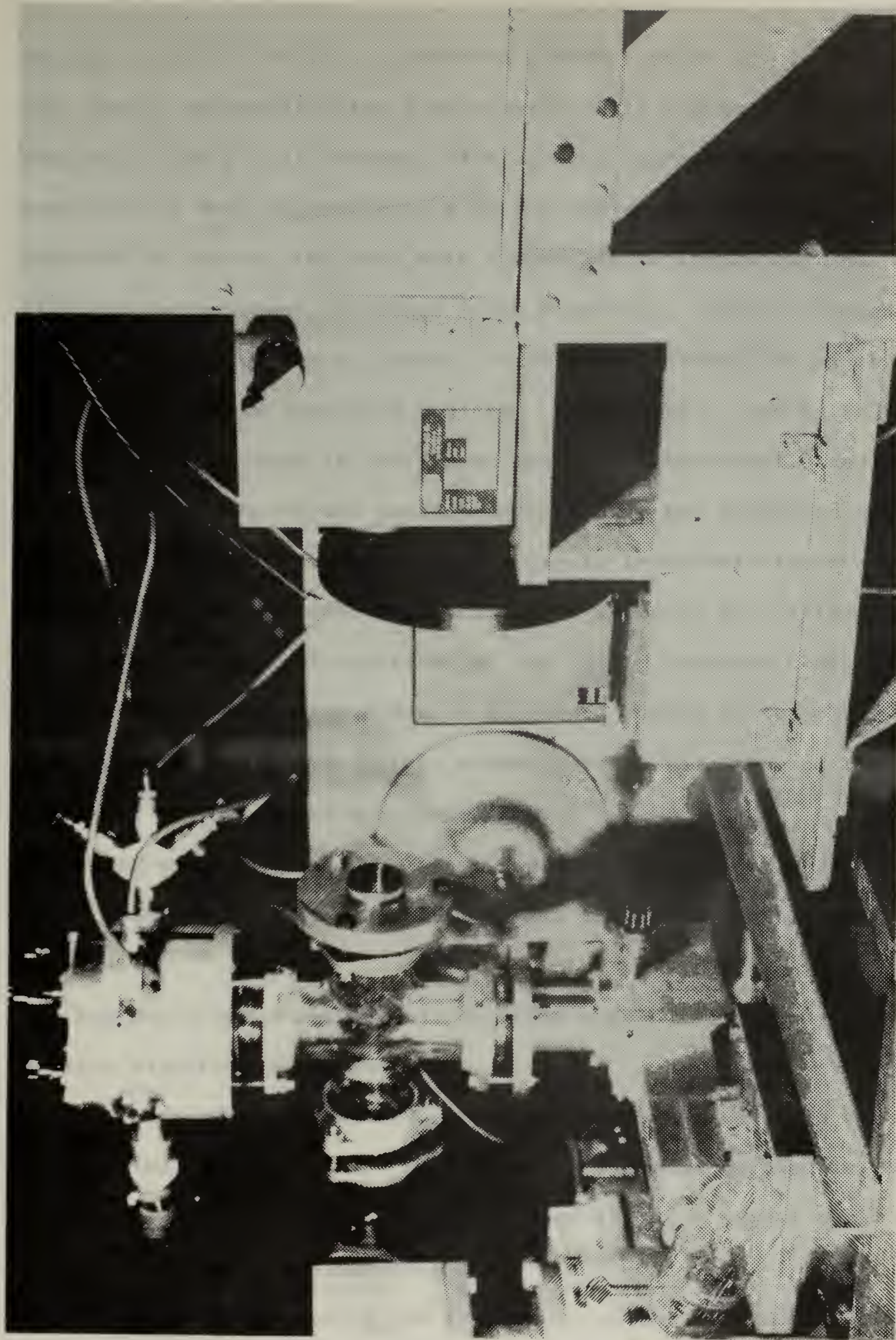


Figure 2. Interaction Chamber and Target Area.

through an exit slit into a modified Electro-Optics Associates type PM-101 photomultiplier tube assembly with a Dumont 6467 PM tube which had a S-11 response with peak at approximately 3500 \AA . Reduction of dark current output of the photomultiplier tube was achieved by cooling the tube with a combination liquid and gas nitrogen system shown in Figure 3 and Figure 4. Liquid nitrogen was passed through copper cooling coils formed around the photo tube envelope inside an insulated housing. The liquid N_2 was collected in a Dewar flask when it overflowed from the tube assembly and the boil off of N_2 vapors was introduced back into the insulated housing to prevent formation of condensate. Liquid level was electrically controlled using a thermocouple sensor to initiate the filling of the system and control operation of the liquid nitrogen control valve. Condensate was avoided on the front window of the photo tube by using a evacuated glass window between the tube and the exit slit of the Jerral Ash monochrometer.

Electrostatic shielding was accomplished by enclosing the photo tube and cooling assembly inside a light tight steel enclosure.

The phototube envelope was placed in a Mu-metal cylinder to achieve electromagnetic shielding.

The monochrometer was aligned to the desired wavelength by sending the output of the P.M. tube to a Princeton Applied Research lock-in amplifier, Model H-8, so the spectral line signal could be peaked prior to an experimental run.

During an experimental run the output of the PM tube was sent to an Ortec preamplifier, model 101; a Canberra amplifier, model

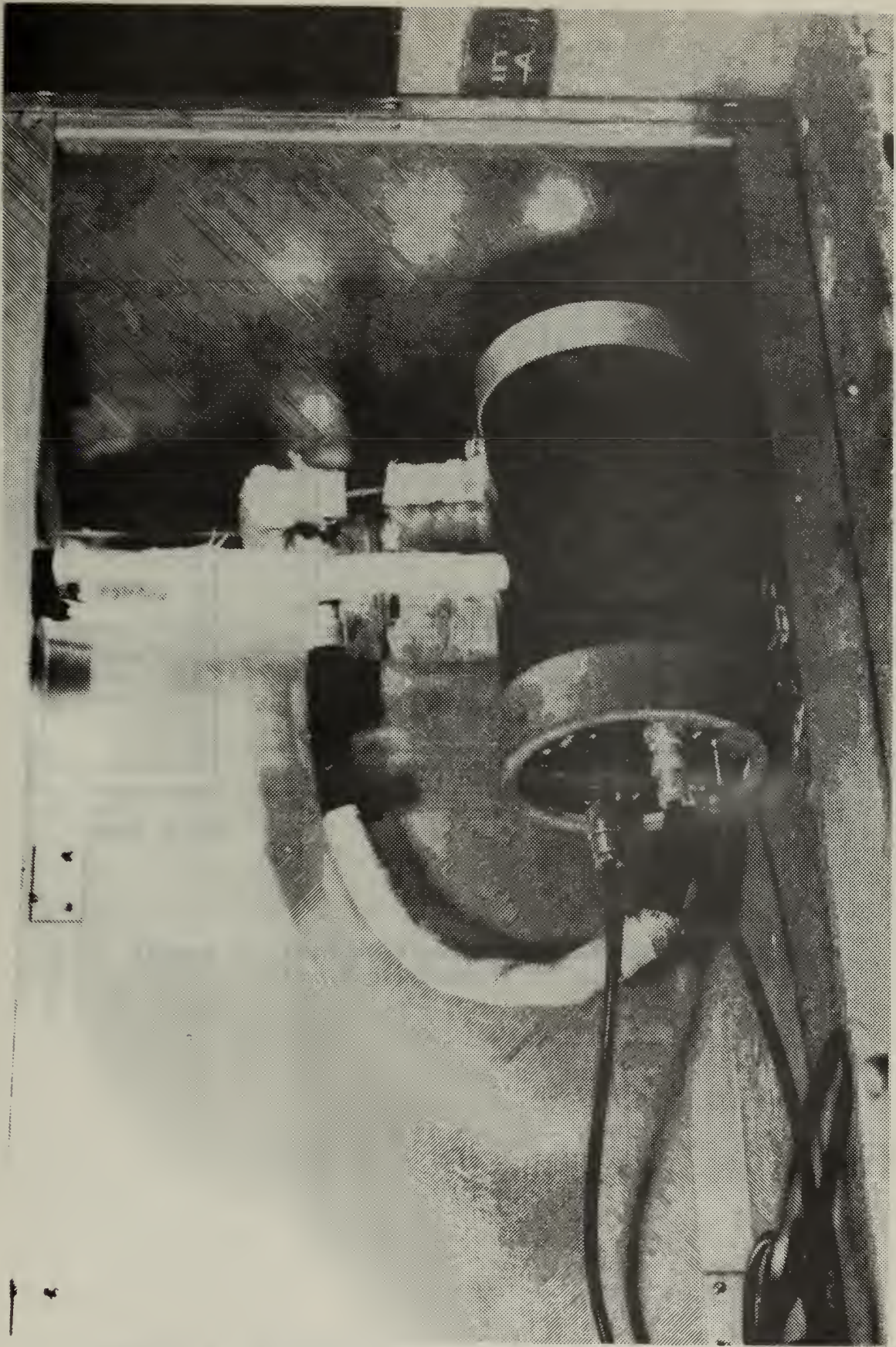


Figure 3. Photomultiplier Tube and Cooling Assembly.

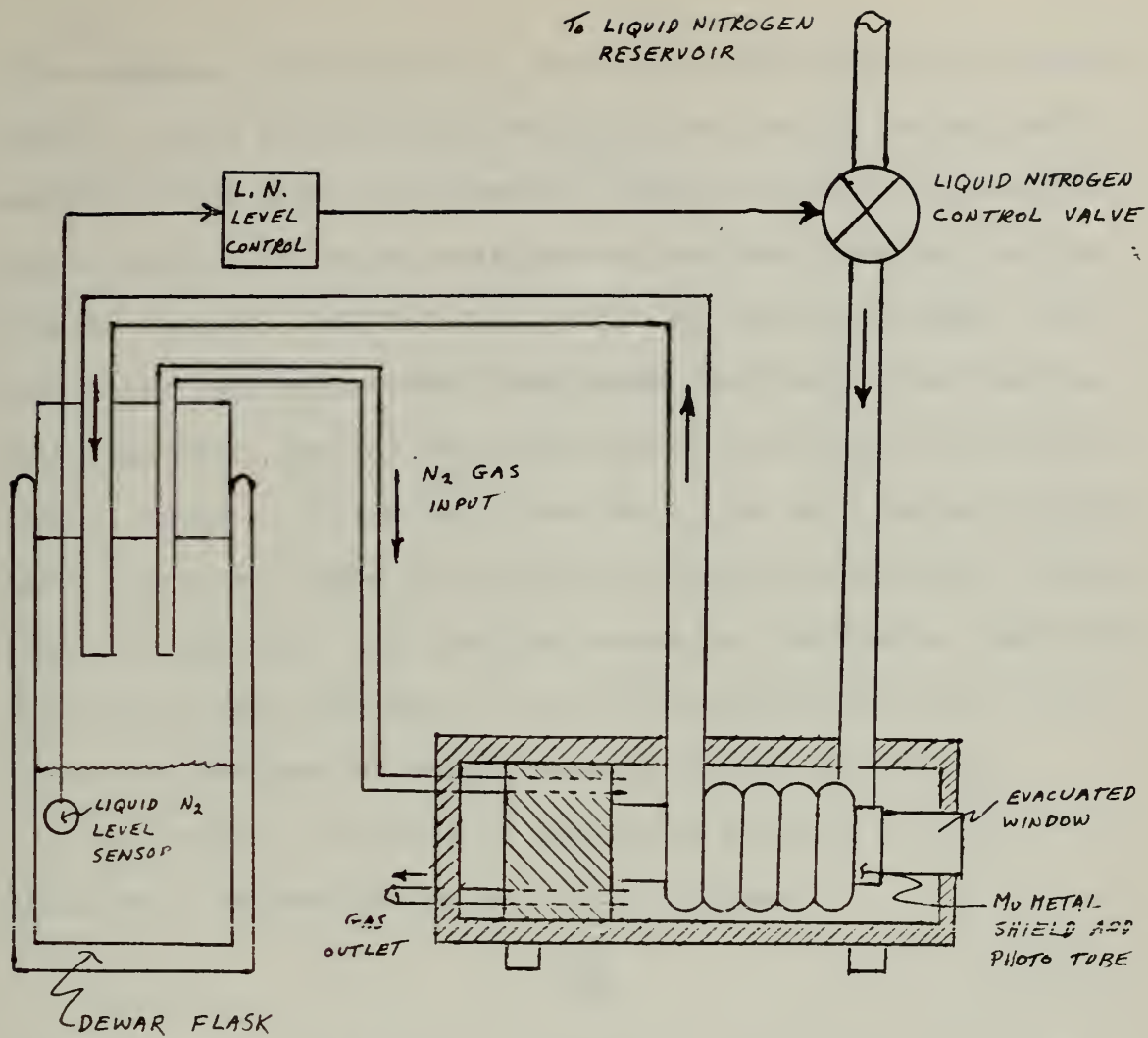


Figure 4. Photomultiplier Tube Liquid/Gas Nitrogen Cooling System.

810; a Canberra discriminator, model 830; and then to two Canberra scalers. Both scalers were controlled by a gating network which enabled pulses from the mechanical chopper at 78 Hz to gate one scaler during the period that the PM tube was receiving photons from the target, and the other scaler during dark periods. Thus one scaler counted photons, background, and dark current pulses while the other counted only dark current and background pulses. Another Canberra scaler was driven at 0.1 Hz by a Berkeley double pulse generator, model 903, and was utilized to determine counting time for each run. The counting system was enabled by a gate from the beam current integrator so that the system was active from 0 to 20 microcoulombs of charge received at the faraday cup.

The relative intensity of the photon emission from the interaction chamber was calculated as follows:

$$I = \frac{NC}{QB} . \quad (1)$$

In equation (1), QB is the total proton beam charge collected, and NC is the total corrected photon count. The total corrected photon count is obtained by subtracting the outputs of the scalers after correcting for dead time.

$$NC = N_1 - N_2 \quad (2)$$

where N_1 is the count of the scaler recording the photon, background and dark current with dead time correction. N_2 is the count of the scaler recording dark current and background with dead time correction. The dead time correction applied to the scaler count was calculated using a dead time (τ) of 2 microseconds as determined by oscilloscope measurements.

$$N(1,2) = \frac{N'(1,2)}{\frac{1-2N'\tau}{T}} \quad (3)$$

where τ is the system dead time, T is the counting period, and N' is the scaler count recorded during an experimental run.

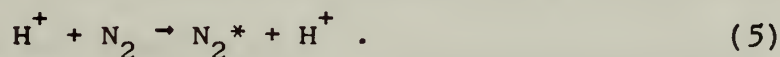
The spectral intensity is independent of the beam current and is a function of the number and energy of the protons and the pressure and temperature of the target gases. The proton beam energy was constant during the runs. The temperature of the target gases is assumed to be a constant during the run. The value of QB was a constant 20 microcoulombs of charge.

The entrance and exit slits were perpendicular to the proton beam and the monochrometer was positional so that the photons generated in the first millimeter of beam in the interaction chamber were focused on the slits. In this region, the energy loss by the protons is small and pressure effects on proton beam energy are minimized.

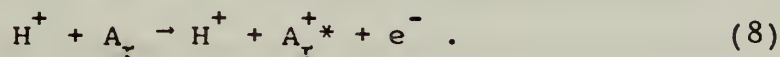
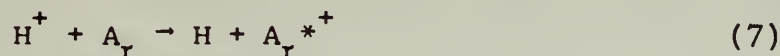
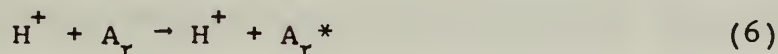
III. THEORY

A. BACKGROUND

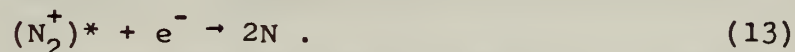
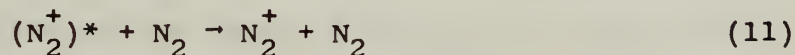
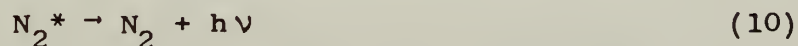
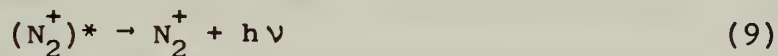
The primary nitrogen reactions occurring in the interaction chamber as a result of proton (H^+) bombardment for the lines studied are:



The primary argon excitation reactions expected to occur are:



The primary de-excitation reactions of the nitrogen first negative and second positive systems are:



The excitation and de-excitation of the nitrogen systems with argon are given in a later section.

The general rate equation [3] was used to develop a set of equations from which the excitational rate could be computed.

$$\frac{dN_k^*}{dt} = PVN\sigma_k + \sum_{l>k} \lambda_{lk} N_l^* - \sum_{k>i} \lambda_{ki} N_k^* - \sum_j N_k^* K_j \eta_j + \sum_m \sum_n \eta_m^* K_{mn} N_n \quad (14)$$

The first term ($PVN\sigma_k$) is the direct excitation to state K by protons in terms of the excitation cross section σ_k , proton beam density P, target density N, and proton velocity V. The second term ($\lambda_{lk} N_l^*$) is the contribution to state K from higher excited states l in terms of the transition probability λ_{lk} from state l into state K, and the number density of excited particles N_l^* in state l. The third term ($\lambda_{ki} N_k^*$) is the radiative loss from state K by transition probability λ_{ki} , and the number density of particles in state k. The fourth term ($N_k^* K_j \eta_j$) is the collisional de-excitation rate K_j due to collisions between particles of target gas in state K with another type of gas particle η in state j. The fifth term ($\eta_m^* K_{mn} N_n$) is the excitation of target molecules to state K by collisions with η type molecules in excited state η_m^* , in terms of the density of particles in excited state η_m^* , the collisional excitation rate K_{mn} , and the density of target molecules in state N_n . This general equation is applicable when investigating a mixture of two gases as well as for each of the constituent gases in the mixture. Thus ($\frac{dN_k^*}{dt}$) the rate of transition from a particular excited state is of this form for both the Argon and the Nitrogen, and the rate equations are coupled in the fourth and fifth terms.

The energy dependence of cross sections in the rate equation necessitated holding the incident proton beam energy constant during the experiment so that the cross sections could be

considered constant for a given transition. Each experimental data point was obtained with constant proton beam density P , velocity V , and gas density constant N . Therefore the first term is a function only of target gas pressure in the interaction chamber.

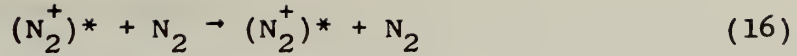
$$P V N \sigma_k = R_k(P). \quad (15)$$

In the second term, states above K can be populated by direct interaction with the incident proton beam. These higher lying states can then decay to state K . This process is assumed to be proportional to the target gas pressure in the interaction chamber also and can be considered additive to the first term to simplify the rate equation.

In the third and fourth term it is assumed that the transition probabilities are constant for a given spectral line and that the number density of target gas particles N_k^* is proportional to the measured photo current intensity (I). In addition, the number density of constituent gas particles of type j is proportional to the partial pressure of the type j particles. This assumption follows from the ideal gas law, which is valid in the pressure ranges used in this study [4].

The fifth term represents the excitation of target gas molecules in state n to state η_n^* by collision with constituent gas particles η_m^* in state m . For proton bombardment on molecular nitrogen alone, the excited species N_m^* will be either $(N_2^+)^*$ or N_2^* depending on which line is under observation. The unexcited species N_n will be N_2 in both cases. Thus the possible collision

mechanism will be:



In either reaction there is no net change and the term can be neglected. The fifth term applies to those excited species having kinetic energies with a Boltzman distribution which is assumed to hold in the reaction chamber.

Under conditions of equilibrium the excitation to state K is equal to the de-excitation from state K, and after applying the above assumptions, the general rate equation becomes:

$$\frac{dN_k^*}{dt} = 0 = R_k - \lambda N_k^* - \eta_j^{K_j} N_k^* + \eta_m^* K_{mn} N_n \quad (18)$$

if

$$N_k^* = bI, R_k = aP, P_j = \eta_j^{K_j} kT, \eta_m^* K_{mn} N_n = f(P_A P_j) \quad (19)$$

$$aP_j = \lambda bI + \frac{K_j b}{kT} P_j I - f(P_A P_j) \quad (20)$$

$$P = \frac{\lambda b}{a} I + \frac{K_j b}{akT} PI - \frac{1}{a} f(P_A P) \quad (21)$$

let

$$A = \frac{\lambda b}{a}, B = \frac{K_j b}{akT}, \quad (22)$$

$$P = AI + BPI - \frac{1}{a} f(P_A P) \quad (23)$$

$$I = \frac{P + \frac{1}{a} f(P_A P)}{A + BP} . \quad (24)$$

B. NITROGEN AS THE TARGET GAS

The rate equation becomes

$$I = \frac{P}{A+BP} \quad \text{since } P_A = 0 \quad (25)$$

thus
$$\frac{P}{I} = A+BP. \quad (26)$$

By plotting $\frac{P}{I}$ versus P , a straight line should result with the intercept giving the value of A and the slope giving the value of B . In this experiment the constants A and B were solved for using this technique.

By taking the ratio of B to A the reaction rate, K_j , can be determined.

$$K = \frac{BKT}{A\tau} \quad (27)$$

the radiative lifetime, τ , is the reciprocal of the decay probability λ . The following experimentally determined values of radiative lifetimes for nitrogen by Bennet and Dalby [5] were used in this experiment.

$$\tau = 6.58 \times 10^{-8} \text{ sec.} \quad 3914\text{\AA} \text{ first negative} \quad (28)$$

$$\tau = 4.45 \times 10^{-8} \text{ sec.} \quad 3371\text{\AA} \text{ second negative} \quad (29)$$

C. NITROGEN WITH ARGON

The experimental results obtained using mixtures of Nitrogen and Argon, Figures 5-11, indicate that the Argon tends to enhance the $(N_2)^*$ and $(N_2^+)^*$ states significantly and the straight line function justifies neglecting the collisional de-excitation term for which the intensity has a $\frac{1}{P_A}$ dependence.

The rate equation (19) can be written for the Argon constituent as:

$$\frac{dN_k^*}{dt} = A_a P_a - \lambda_a N_k^* - k N_k^* N_k + K_{mn} N_k^* N_n \quad (30)$$

here, again, the second term on the right hand side represents population of an argon excited state by higher states and the third term represents the depopulation by transition from the excited state of interest to lower excited states.

Equations (30) and (18) are related by the fourth term. The argon rate equation in equilibrium results in

$$K_{mn} N_k^* N_n = P_a - \lambda_a N_k^* - K_j N_k^* \eta_j. \quad (31)$$

Now if $N_k^* = b' I$ with other terms defined by equation (20).

$$K_{mn} N_a^* N_n = A_a P_a + \lambda_a b' I + \frac{K_j b'}{kT} P_A I. \quad (32)$$

If the energy transfer from argon to nitrogen is much greater than the rate of self collisional de-excitation then the second and third terms can be neglected and equation (32) reduces to

$$K_{mn} N_a^* N_n \cong P_a A_a. \quad (33)$$

This approximation holds for the first negative transition ($\lambda = 3914.4\text{\AA}$) so that for this case equation (24) can be written as:

$$I_1 = \frac{P+C' P_a}{A+B P} \quad (34)$$

$$\text{where } C' = \frac{A_a}{a}. \quad (35)$$

If the frequency of an argon transition is the same as the transition of interest as in the case of the second positive transition ($\lambda = 3371.3\text{\AA}$) then

$$K_{mn} N_k^* N_n = A_a P_a - \lambda_a N_a^* - k N_a^* N_a \quad (36)$$

$$K_{mn} N_k^* N_n = A_a P_a - \lambda_a b' I - \frac{k_j b'}{kT} P_a I \quad (37)$$

then from equation (18)

$$0 = R_k - \lambda N_k^* - \eta_j K_j N_k^* + A_a P_a - \lambda_a b' I - \frac{k_j b'}{kT} P_a I \quad (38)$$

$$0 = aP - \lambda bI - \frac{K_j b}{kT} PI + A_a P_a - \lambda_a b' I - \frac{k_j b'}{kT} P_a I \quad (39)$$

let

$$B' = \frac{k_j b'}{akT}, \quad A' = \frac{\lambda_a b'}{a}, \quad C' = \frac{A_a}{a} \quad (40)$$

$$0 = P - AI - BPI + C' P_a - A' I - B' P_a I \quad (41)$$

$$(A + BP + A' + B' P_a) I = P + C' P_a \quad (42)$$

$$I_2 = \frac{P + C' P_a}{A + BP + A' + B' P_a} \quad (43)$$

If intensity is plotted as a function of total pressure, and letting $I_N = \frac{P}{A + BP}$ = the intensity of radiation at maximum N_2 pressure before the addition of argon, then equation (34) can be written as:

$$I_1 = I_N + \frac{C' I_N}{P} P_a \quad (44)$$

and the slope of the straight line function is the quantity

$S = \frac{C'}{P} I_N P_a > 0$. Where S_1 is the slope of the line while P is

the maximum pressure of nitrogen. Then

$$C' = \frac{PS_1}{I_N} \quad (45)$$

here C' is the excitation coefficient for the first negative transition and is a function of $C' = C' (P_i, V_i, N_i, \sigma_i)$.

Using the same arguments for the second positive transition ($\lambda = 3576\text{\AA}$) the value of the second positive excitation coefficient can be determined. Then writing

$$I_2 = \frac{P + C' P_a}{A + BP + A' (1 + \frac{B'}{A'}) P_a} \quad (46)$$

$$I_2 = \frac{1 + C' \frac{P_a}{P}}{I_N + \frac{A'}{P} (1 + \frac{B'}{A'} P_a)} \quad (47)$$

The ratio $\frac{B}{A}$ is found in [6] and all other constants are known with the exception of A' . If equation (48) is solved for A' then:

$$A' = \frac{P}{(1 + \frac{B'}{A'} P_a)} \left[\frac{1 + C' \frac{P_a}{P}}{I_2} - I_N \right] \quad (48)$$

If this value of A' is then determined experimentally for the second positive transition ($\lambda = 3371.3\text{\AA}$) and substituted back into equation (47) then the theoretical value of I_2 can be calculated and compared with the experimental results.

IV. RESULTS

A. GENERAL

The experimental determination of the Intensity versus Pressure relationship are shown by Figures 5-11 for the first negative ($\lambda = 3914.4\text{\AA}$) and second positive ($\lambda = 3576\text{\AA}$ and $\lambda = 3371.3\text{\AA}$) transitions. The value of excitation coefficient was determined for the first negative transition using equation (45) and found to be $.256 \pm .046$. The value excitation coefficient for the second positive transition was determined for ($\lambda = 3576\text{\AA}$) using the data shown in Figure 8 and found to be $2.25 \pm .12$. This value was then verified by a comparison of the theoretical computation using equation (47) and comparing this with the experimental results found in Figures 10 and 11 for various pressures of Nitrogen.

B. DISCUSSION

The assumptions made in arriving at equation (26) were verified by comparison and close correlation between experimental results and theoretical equations [2]. Secondary excitation effects, such as the excitation by high energy electrons and soft x-rays, were neglected in arriving at equation (26). It was assumed that the only excitation processes were direct, and that this was proportional to the pressure of the target gas.

In developing the pressure versus intensity relationship for the nitrogen-argon mixtures, it was assumed that the de-excitation of the nitrogen due to collisions with argon was negligible. This seems valid since it would result in a $1/P$ dependence instead of a linear function which is found experimentally.

Some nonlinearity is found in the intensity versus pressure curves at total pressure values of greater than 500 torr. This is believed to be a result of the energy loss which the protons experience while traveling in the interaction chamber and the finite length of beam which is viewed by the instrumentation. Thus at higher pressures the assumption that the beam is monoenergetic starts to fail.

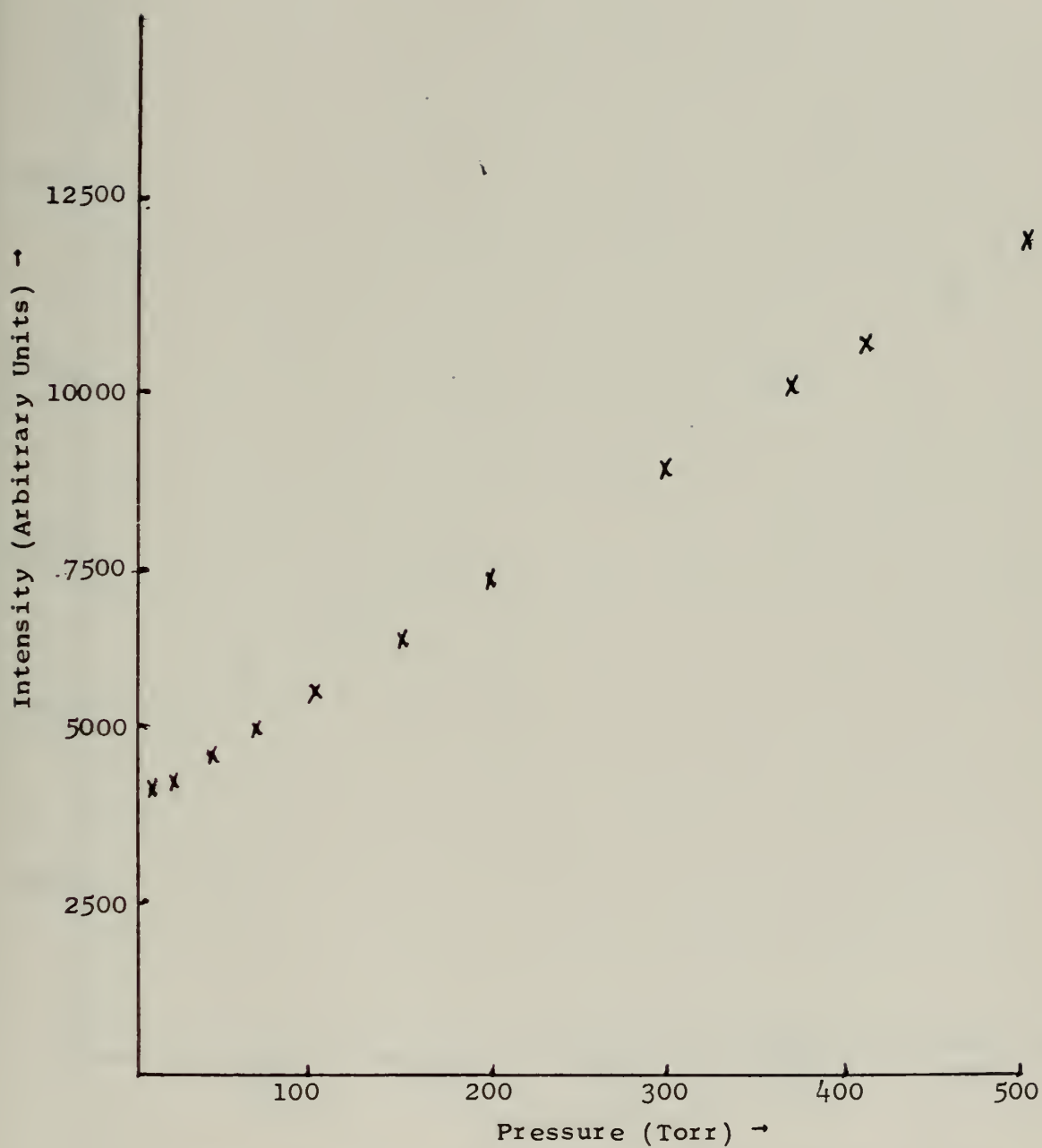


Figure 5. Intensity versus Pressure (3914\AA). H^+ on 10 Torr N_2 , Balance A. Experimental Data.

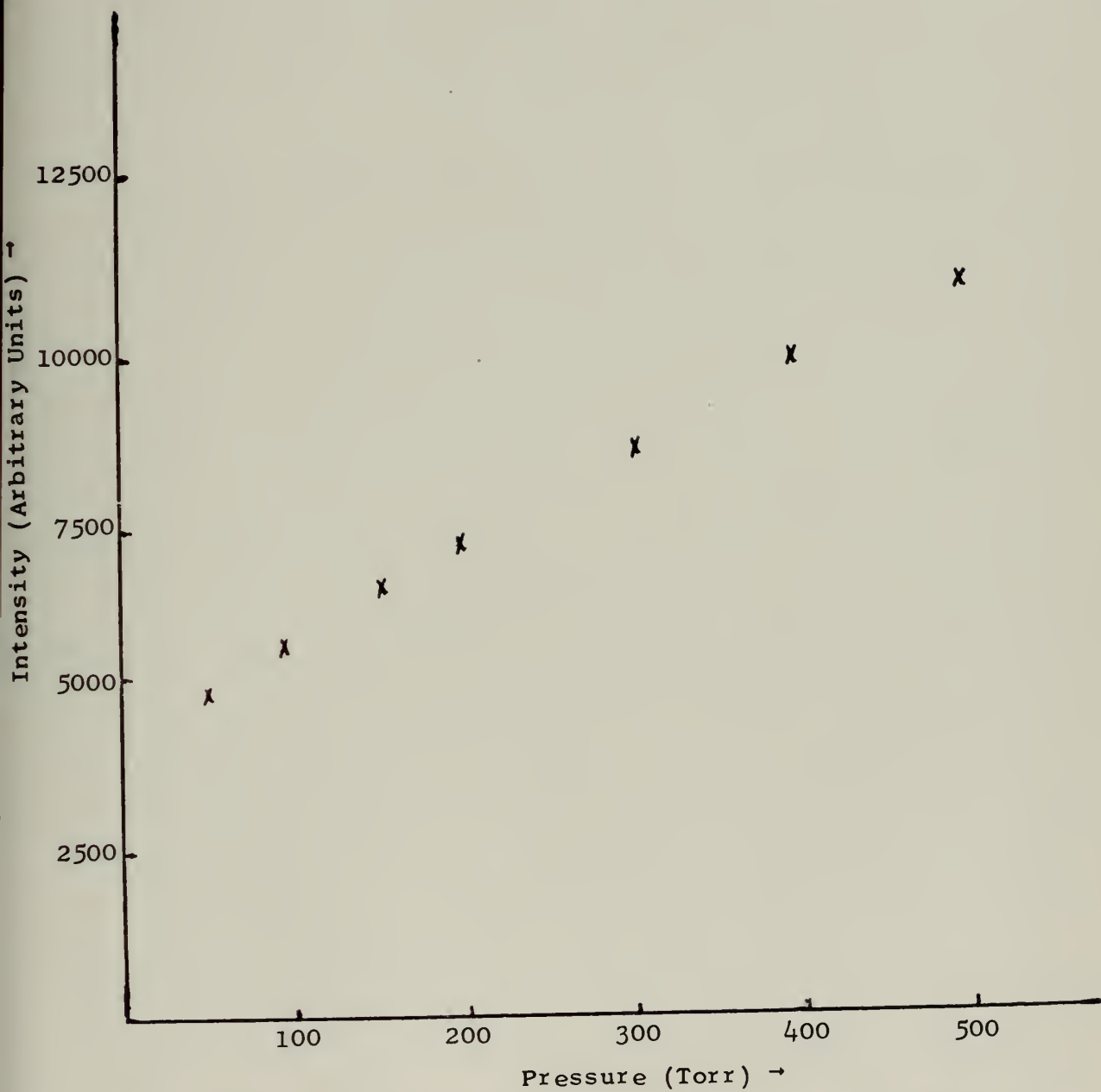


Figure 6. Intensity versus Pressure (3914\AA). H^+ on 50 Torr N_2 , Balance A. Experimental Data.

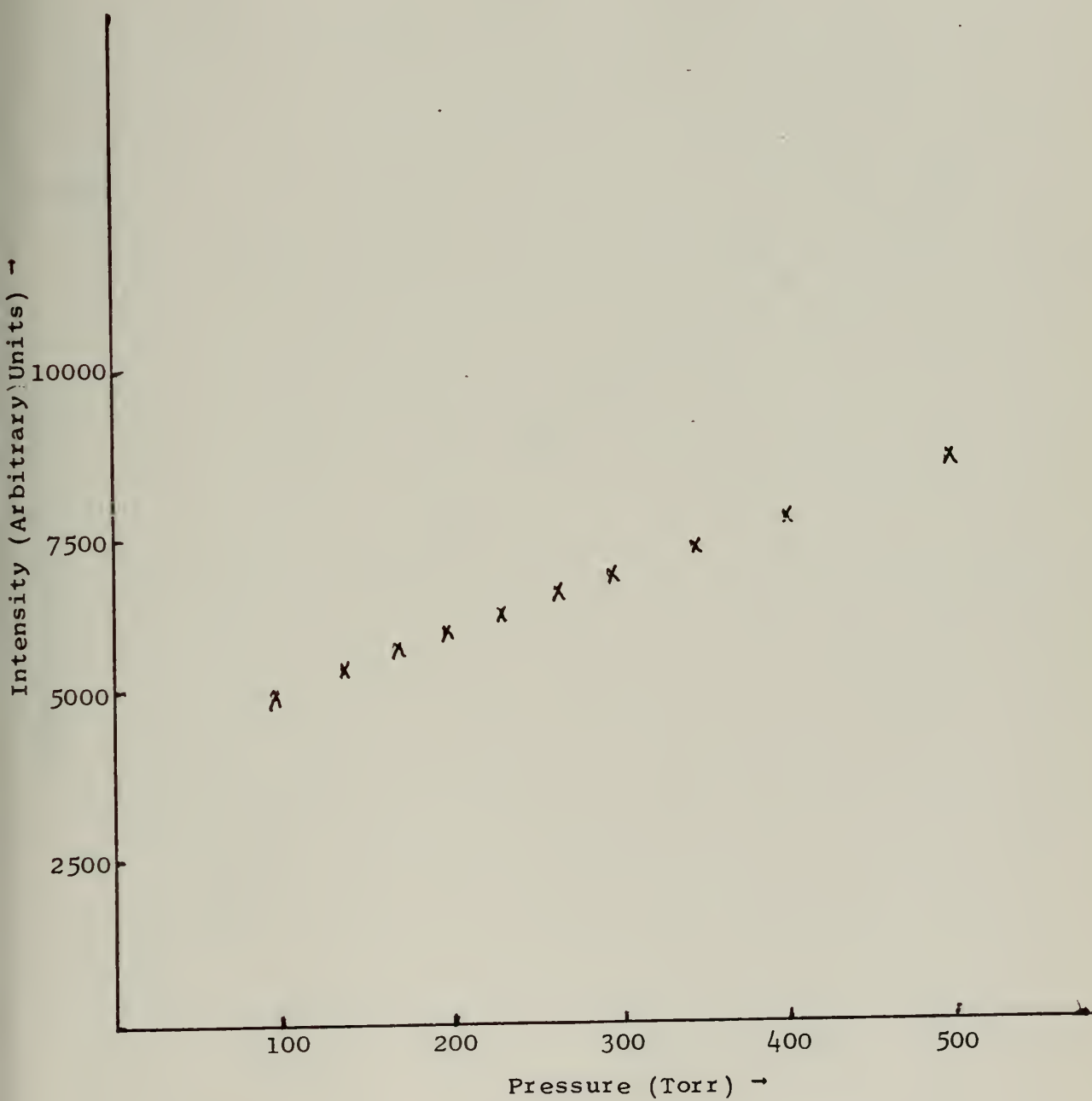


Figure 7. Intensity versus Pressure (3914\AA). H^+ on 100 Torr N_2 , Balance A. Experimental Data.

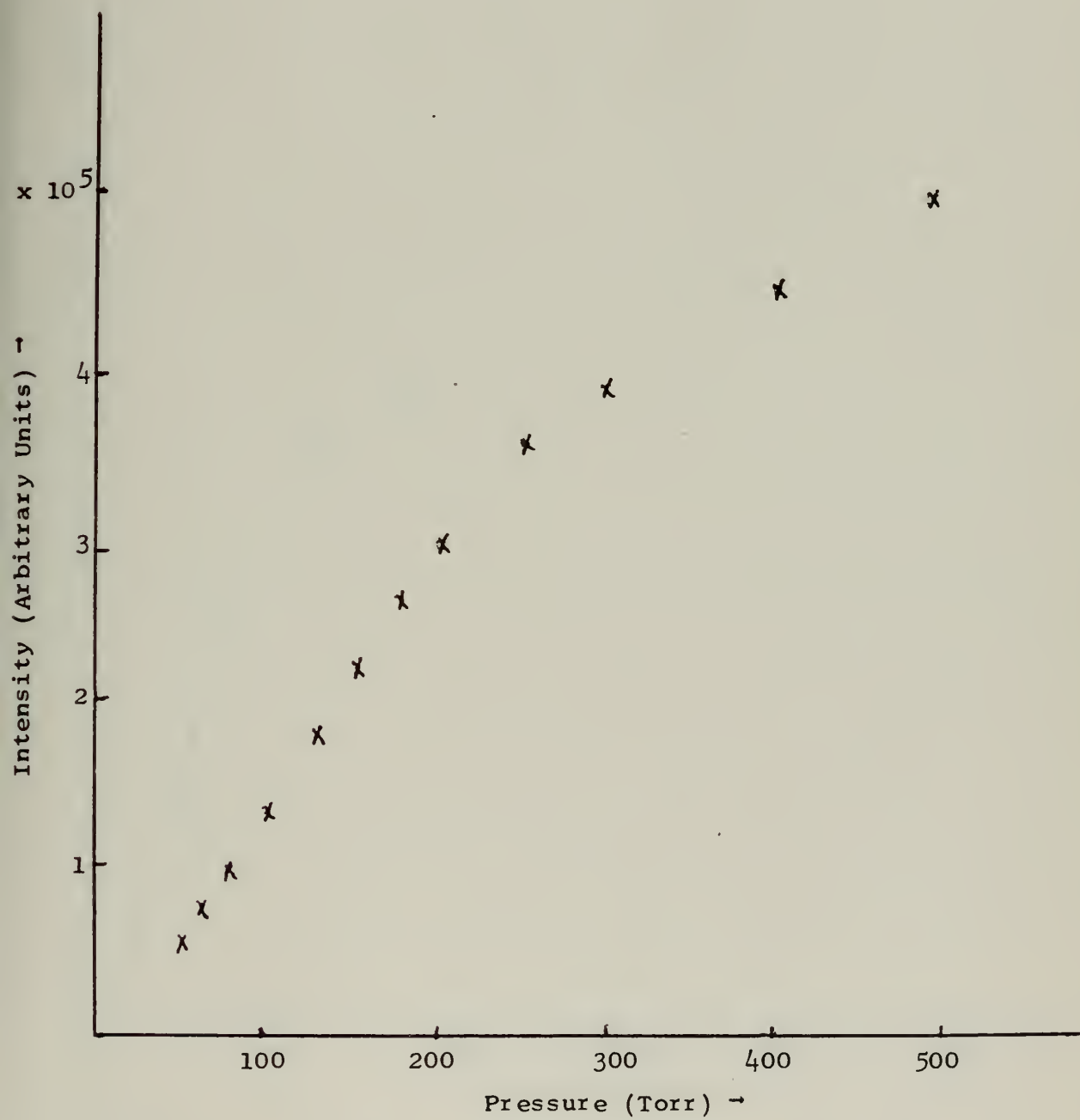


Figure 8. Intensity versus Pressure (3576\AA) H^+ on 50 Torr N_2 , Balance A, Experimental Data.

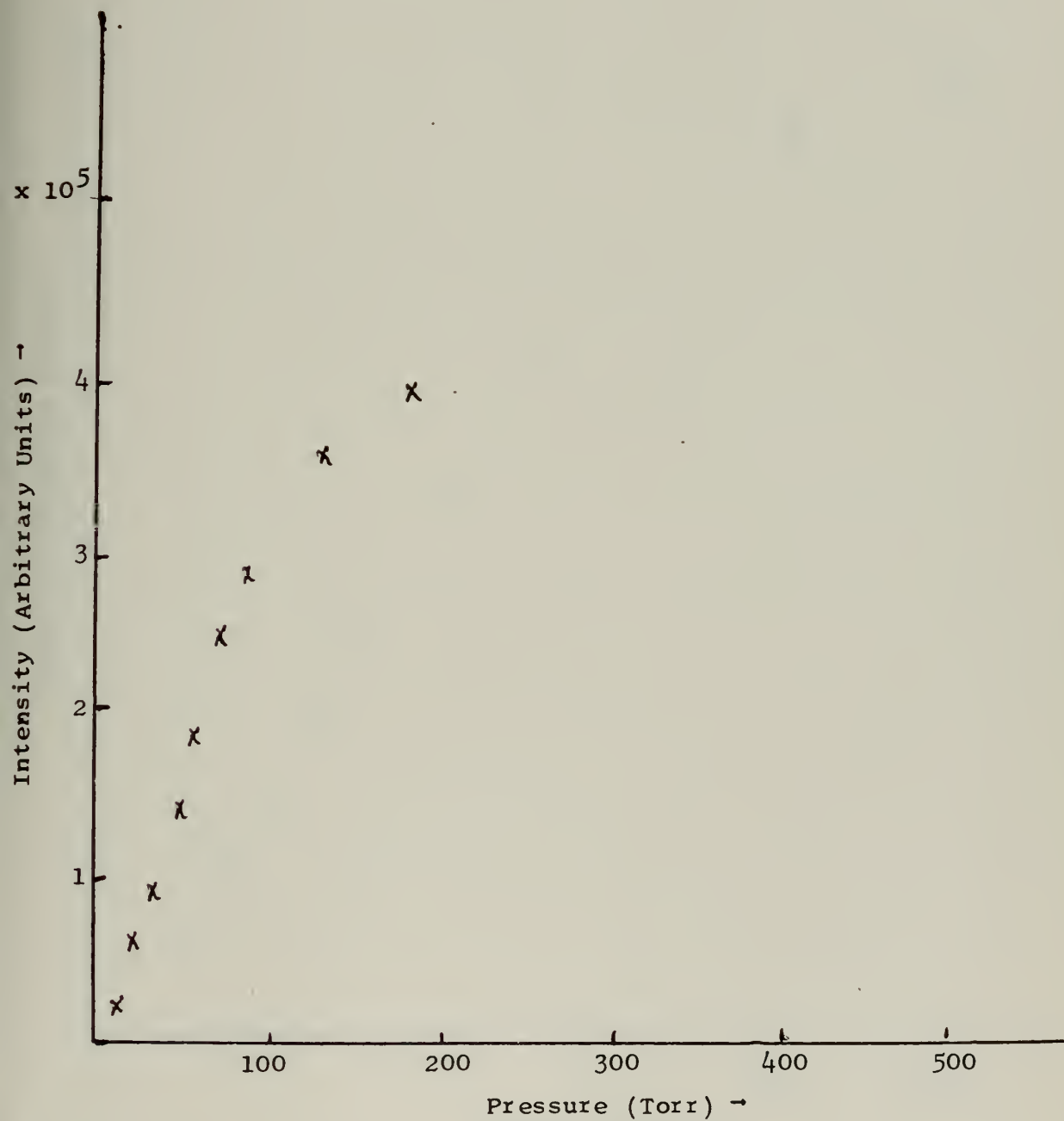


Figure 9. Intensity versus Pressure (3371\AA). H^+ on 10 Torr N_2 , Balance A. Experimental Data.

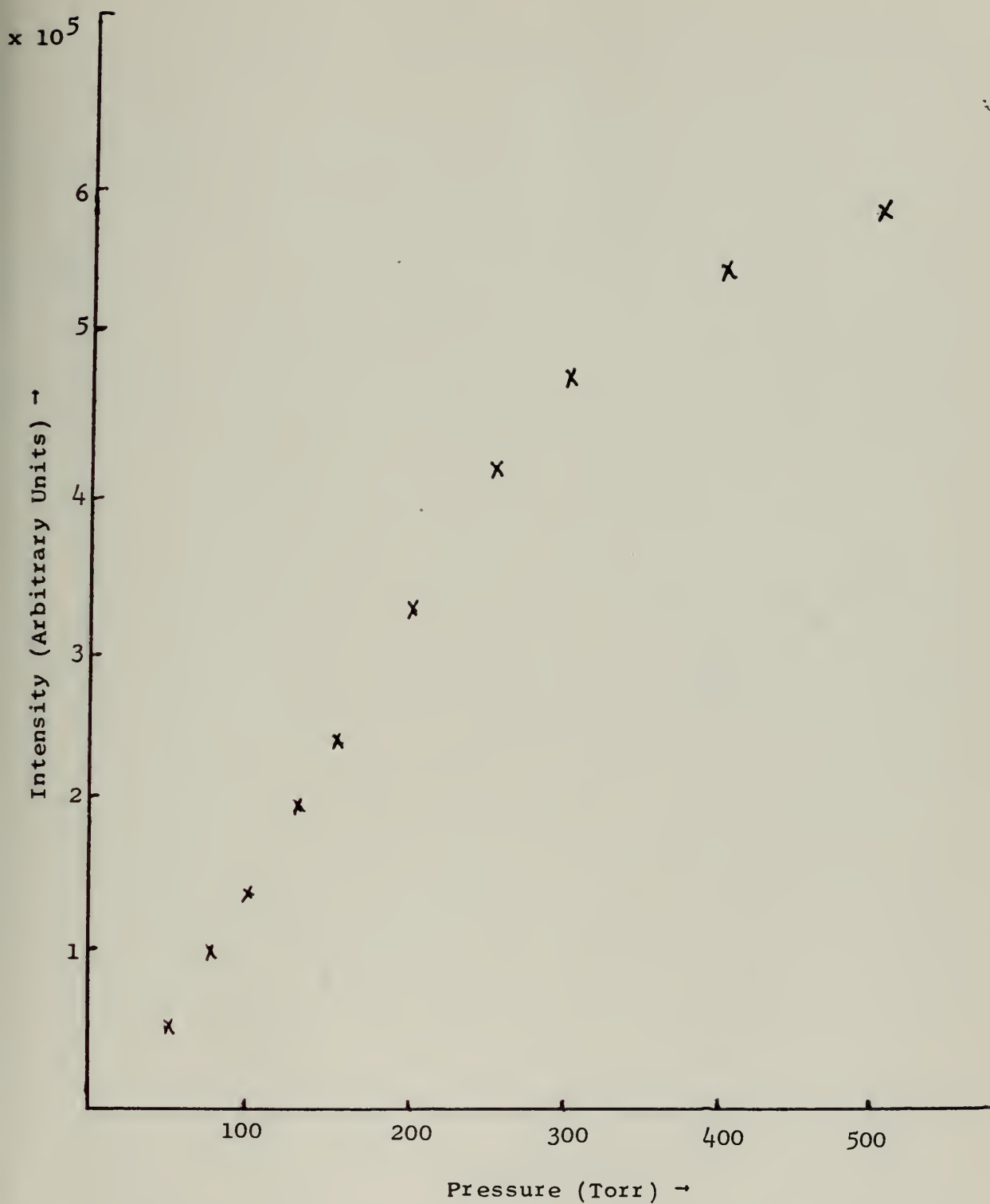


Figure 10. Intensity versus Pressure (3371\AA). H^+ on 50 Torr N_2 , Balance A. Experimental Data.

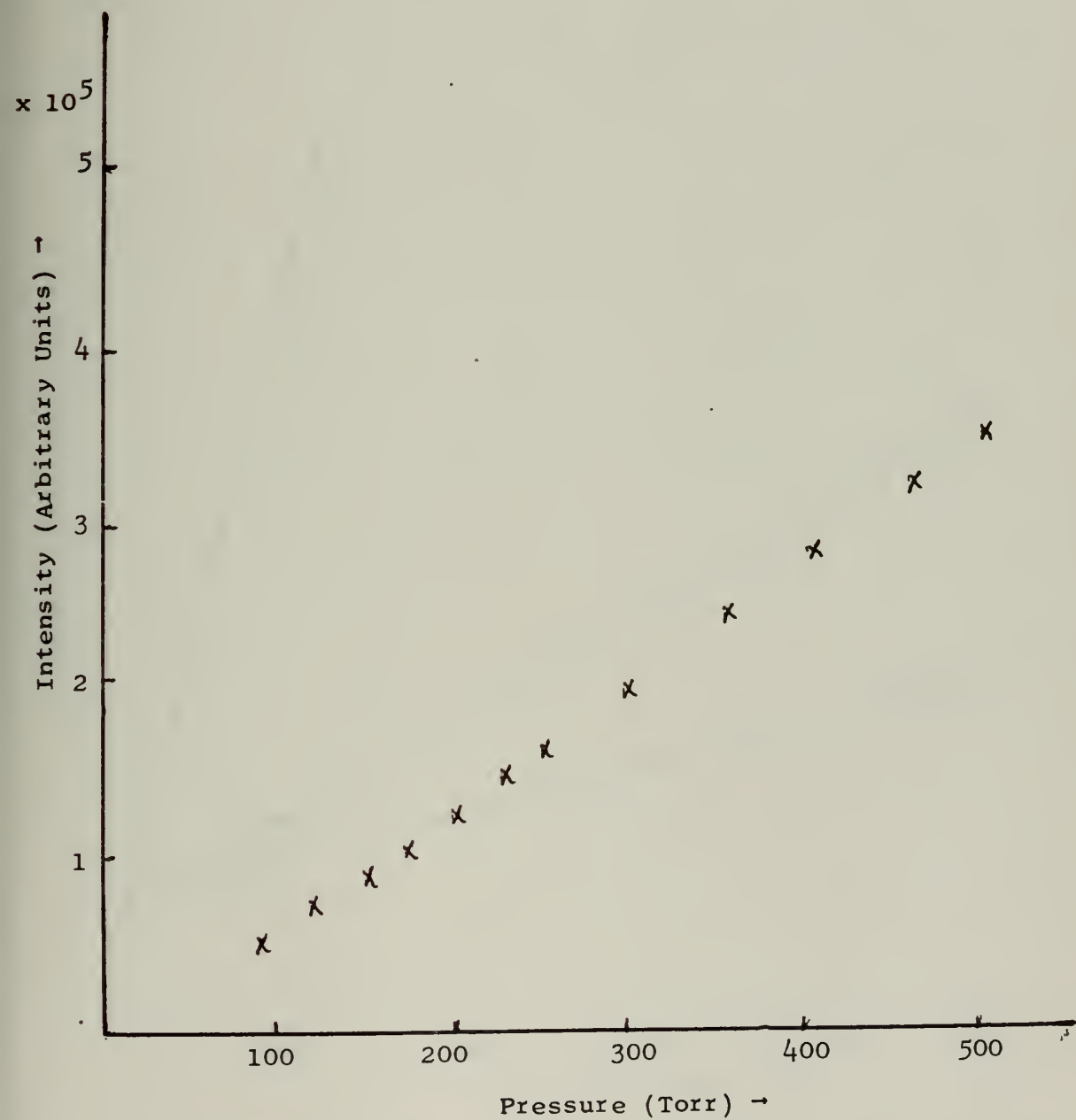


Figure 11. Intensity versus Pressure (3371Å). H^+ on 100 Torr N_2 , Balance A. Experimental Data.

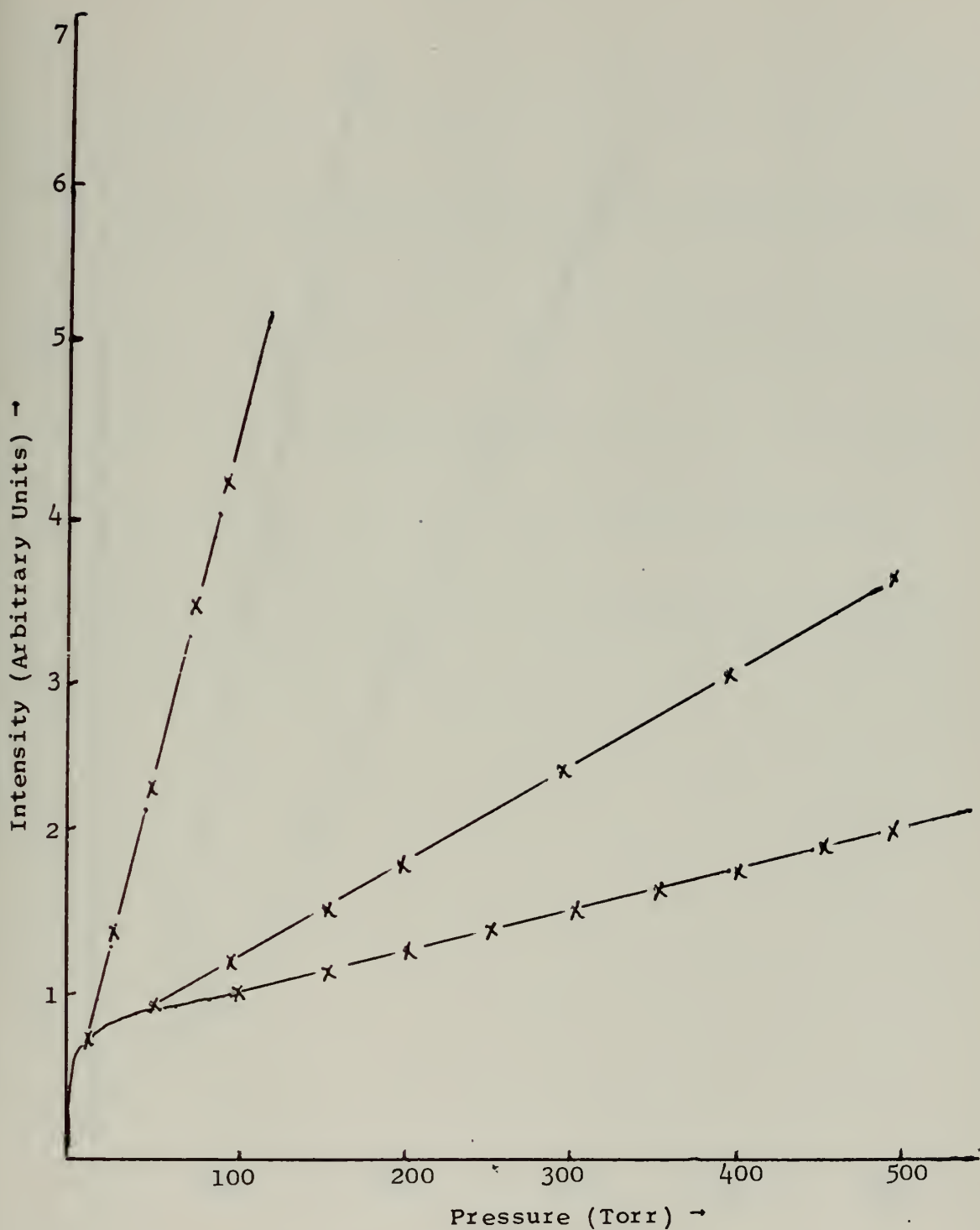


Figure 12. Intensity versus Pressure (3914\AA). H^+ on N_2 at 10, 50, and 100 Torr, Balance A. Normalized to Intensity at 100 Torr N_2 . Experimental Data (X). Theory Comparison.

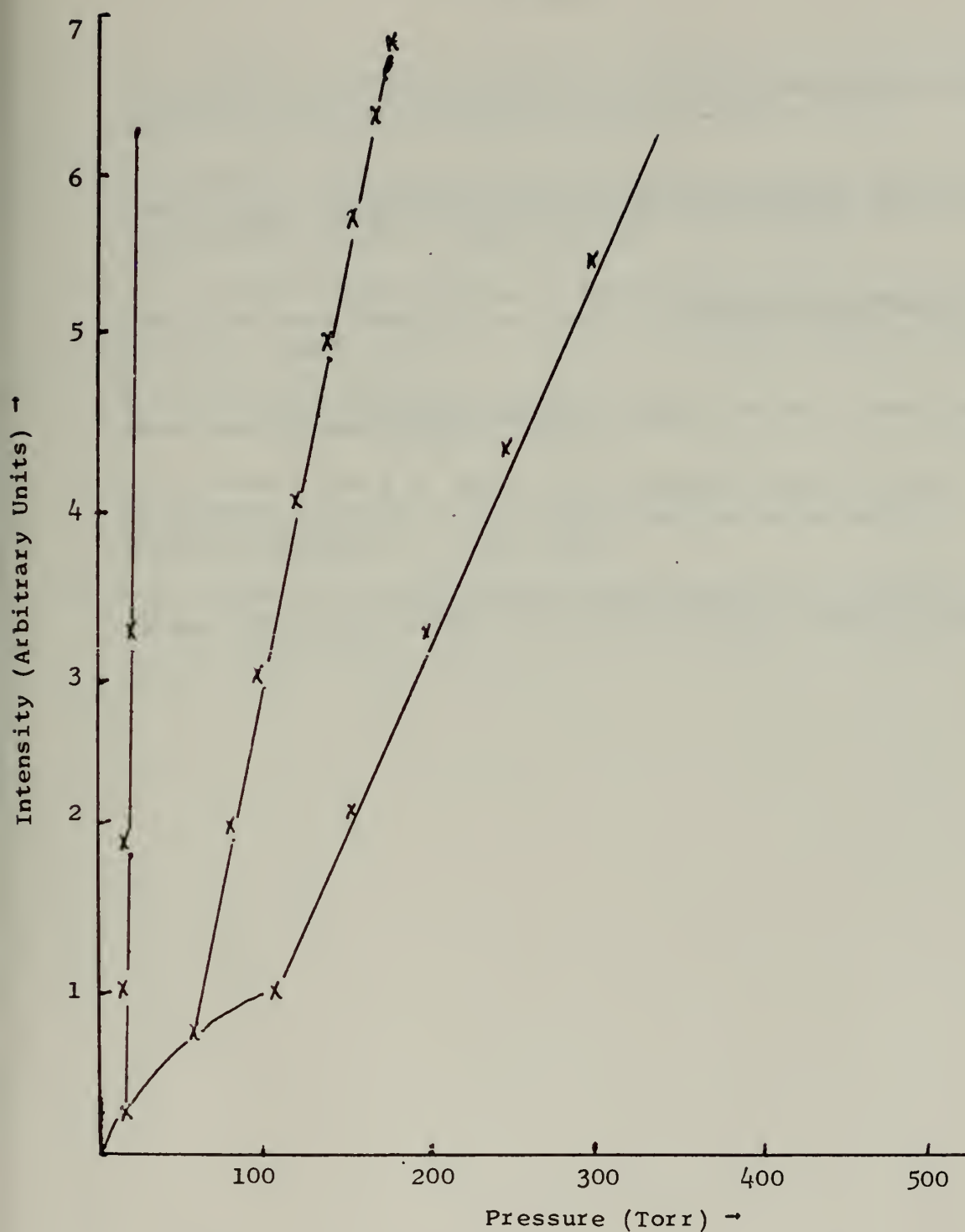


Figure 13. Intensity versus Pressure (3371\AA). H^+ on N_2 at 10, 50, and 100 Torr, Balance A. Normalized to Intensity at 100 Torr N_2 . Experimental Data (X). Theory Comparison.

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13. ABSTRACT Nitrogen and nitrogen - argon gas mixtures were excited by 1.5 MeV protons from a Van De Graaff generator. Intensity measurements were made for the first negative ($\lambda = 3914.4\text{\AA}$) and the second positive ($\lambda = 3371.3\text{\AA}$) transitions of molecular nitrogen. A determination of the excitation coefficient (C), which is a function of the reaction rate coefficient and excitation cross section, was made for the nitrogen first negative and second positive transitions. The excitation coefficient (C) is a constant for each transition and contains all non-radiative energy transfer constants of the general rate equation. For the first negative transition the value of excitation coefficient was found to be $.256 \pm .046$ and for the second positive transition it was found to be $2.25 \pm .12$.			

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